Response to reviewer comments: Effect of volcanic emissions on clouds during the 2008 and 2018 Kilauea degassing events

Katherine H. Breen[1, 2], Donifan Barahona[1], Tianle Yuan[1, 3], Huisheng Bian[1, 3], and Scott C. James[4]

[1]NASA, Goddard Space Flight Center, Greenbelt, MD, USA
[2]Universities Space Research Association, Columbia, MD, USA
[3]Joint Center for Earth Systems Technology. University of Maryland, Baltimore County, MD, USA
[4]Baylor University, Departments of Geosciences and Mechanical Engineering, Waco, TX, USA

Correspondence: Donifan Barahona (donifan.o.barahona@nasa.gov)

Review Reports

Christoph Kern:

We thank Dr. Kern for taking the time to comment on the manuscript.

Comment: *This is an important contribution on the interaction of Kilauea volcanic gas and aerosol emissions with meteorological clouds. In reading through the manuscript, I was left with some questions regarding the SO2 emissions data used in this study. The 2008 and 2018 degassing episodes discussed here differed in two ways that I believe may be important for this discussion. For one, SO2 degassing in 2008 occurred mostly at the summit of the volcano while the majority of degassing in 2018 occurred at lower elevations in the lower East Rift Zone. Also, and perhaps more importantly, the SO2 emission rate during May-July 2018 was approximately an order of magnitude greater than during 2008. In both cases, I believe the authors have not yet considered the state-of-the-art in our understanding of SO2 degassing to the atmosphere during these eruptive episodes. Below, I've listed a few more details in this regard which I hope might help the authors to further improve their study.*
Response: Thank you. We have added the following text to the "Experimental configuration" subsection to clarify assumptions made about source elevation vs. the importance on injection height in this study:

> In this work, we assume that the source elevation of emissions (ERZ vs. summit) is irrelevant during peak emissions, but that the injection height of aerosols directly into the troposphere at different altitudes (below or above boundary layer processes) influenced cloud microphysics and macrophysical characteristics for liquid and ice clouds. We also assume the primary aerosol to be SO_2 with some percentage of ash, although it is likely that sea salt contributed to ACIs in liquid clouds and is recommended for inclusion in future parameterizations.

Comment: *The manuscript (e.g. Figure 1 caption) mentions peak sulfate emissions of 50 kt/d. This is confusing in several ways – for one, we (the USGS) did not measure sulfate, but rather SO2 emissions. High temperature volcanic vents like those at Kilauea emit sulfur mostly in the form of SO2. The SO2 is then converted to sulfate over the course of hours to days. Throughout the manuscript, it is therefore probably best to refer to SO2 emissions rather than sulfate emissions. The 50 kt/d value refers to SO2 and was an estimated minimum value reported by Neal et al. 2019. These emissions occurred mostly from*

the lower East Rift Zone, not the summit crater shown in the image which is also confusing. Since the publication by Neal et al. in 2019, we have made significant further progress in quantifying the gas emissions related to the 2018 eruption of Kilauea. As Allan Lerner points out in his comment, please refer to Kern et al. 2020 for this information. For example, we now know that peak SO2 emissions of more than 100 kt/d appear to have been sustained throughout the month of June and into early July 2018 (Figure 10 in Kern et al. 2020). We also broadly discuss the topics of aerosol formation and pyrocumulus cloud formation over the lower East Rift Zone, as well as the coincident gas emissions from the volcano's summit and middle East Rift Zone, all of which the authors may find useful in refining their work.

Regarding the 2008 emissions, please note that Kilauea Volcano was in a state of eruption at its summit Halema'uma'u Crater during the entire 2008-2018 timeframe, not just in 2008. However, the authors are correct in that the highest SO2 emissions (likely > 10 kt/d) occurred during 2008 (see comment below). I would like to encourage the authors to clarify this somewhat, stating that they are focusing on the first year of the 2008-2018 summit eruption during which the highest SO2 emissions occurred, rather than just referring to a 2008 event. I think this would be important given the fact that emissions averaged about 5 kt/d long after 2008 and continued to have a significant impact on environment and air quality in downwind regions during this entire time. See the following two references on this topic:

Businger S, Huff R, Pattantyus A, Horton KA, Sutton AJ, Elias T, Cherubini T (2015) Observing and Forecasting Vog Dispersion from Kilauea Volcano, Hawaii. Bull Amer Meteor Soc 96:1667–1686. https://doi.org/10.1175/BAMS-D-14-00150.1

Pattantyus AK, Businger S, Howell SG (2018) Review of sulfur dioxide to sulfate aerosol chemistry at Kilauea Volcano, Hawai'i. Atmos Environ 185:262–271. https://doi.org/10.1016/j.atmosenv.2018.04.055

For our best estimates of SO2 emissions during the 2008-2013 period, please refer to our recent data release available here:

Elias, T., Kern, C., Sutton, A.J., and Horton, K., 2020, Sulfur dioxide emission rates from Kilauea Volcano, Hawaii, 2008-2013: U.S. Geological Survey data release, https://doi.org/10.5066/P9K0EZII.

Response: Thank you for pointing this out. We have clarified the volcanic state of Kilauea during the eruption period like so:

> The summit of Kilauea has been in an eruptive state since 2008, and the degassing events of 2008 and 2018 represent brief periods of increased volcanic activity and SO_2 emissions resulting in an optically denser plume relative to passive degassing. Several studies have noted the effects of the plume downwind of Kilauea long after violent eruptions have ceased (Businger et al., 2015; Pattantyus et al., 2018), however this study is concerned with ACIs during peak degassing only, when the effects of aerosols on clouds were strongest.

Comment: *Figure 1A in the above reference provides an overview of SO2 emission rates reported by different authors and using various methods. The estimates vary in magnitude but note that, regardless of the utilized methodology, emissions vastly exceeded the 1,000 t/d level mentioned on page 4, line 17 of the manuscript.*

As for the SO2 emissions in 20108, the authors state on page 6, line 10 that they used daily varying SO2 emission rates for their analyses. However, the reference cited is from 2017, so it's unclear where the data corresponding to the 2018 eruption come from. Assuming they come from an analysis of OMI operational SO2 products, it would be quite important to discuss the uncertainty of these data. As described in Kern et al (2020), we had to go to significant effort to account for complex radiative transfer in and around the gas plumes emitted from Kilauea's lower East Rift Zone when analyzing our ground-based DOAS measurements. Similar corrections are likely needed when analyzing satellite remote sensing observations of these dense gas clouds. As we discuss in the 'Future Work' section of Kern et al 2020, operational satellite products are likely to underestimate the true magnitude of emissions without such corrections. It may therefore be better to use the SO2 emission rates reported in Kern et al 2020 for these analyses (the values are included as a supplement to the paper, along with some measurements of plume height).

Response: We have updated the text to clarify the source of 2018 emissions in the Data section like so:

> Daily emissions for 2018 were obtained from Li et al. (2020). Missing values were replaced with Ozone Mapping and Profiling Suite data (https://so2.gsfc.nasa.gov/) whenever possible, otherwise the nearest real data point was used. Vertical column density data were converted from molecules of SO_2 cm^{-2} to kg sulphur per second (kg S s^{-1}) using the linear relationship shown in (Beirle et al., 2014, Fig. 6).

When addressing this comment, we discovered that May 2018 emissions were left constant during the 2018_1× experiment and therefore repeated the 2018 control simulation using the correct emissions. The correction had only a minor effect on the results and didn't modify our conclusions. This is in part because the effect of aerosol on clouds tends to saturate at high aerosol concentration. Hence even a moderate error in emissions would likely have only a minor impact in our results, particularly for the 2018 event. To maintain consistency with the approach used for the 2008 simulations, we have used OMI data (top down) for 2018 as opposed to the suggested emissions from Kern et al. (2020) (bottom up), which we feel is a more thematic comparison with MODIS observations (also top down).

Finally, it is also not clear whether it is valid to initialization of the model with the same plume heights for the 2008 and 2018 events, given that the 2008 emissions occurred from the summit of the volcano and the 2018 emissions mostly occurred from the lower East Rift Zone. I would encourage the authors to clarify the assumptions made in their study in this regard, and as one of the reviewers also states, discuss the uncertainties associated with these assumptions in more detail.

Thank you for the opportunity to provide feedback on this effort. I look forward to reading the final version of this important manuscript.

Response: Thank you for your response - the feedback has provided us with an opportunity to address a few important assumptions. On the topic of plume height, here we are interested in to top of the plume because it is the maximum altitude at which aerosols are directly injected into the atmospheric column. For the sensitivity experiments, we tested if direct interaction

with aerosols above vs. below the boundary layer had significant effects on cloud processes, specifically ice clouds. In 2018, the simulated and observed ACIs showed more similarities than ice clouds in 2008 - in fact, 2008 ice clouds show little to no anomalous cloud processes. This was an interesting finding during the initial research phase and prompted us to ask - why? We knew that emissions were higher in 2018 than 2008, but it had also been reported that plumes had been observed as high as 8 km during peak 2018 emissions. Intuitively, it makes sense that injecting SO2 and ash directly at high latitudes would increase the concentration of ice nucleating particles originating from ash. From our sensitivity experiments, it appears that this intuitive assumption was, at least in part, correct. We recommend the parameterization of other possible "culprits" for future work.

References

Beirle, S., Hörmann, C., Penning de Vries, M., Dörner, S., Kern, C., and Wagner, T.: Estimating the volcanic emission rate and atmospheric lifetime of SO_2 from space: a case study for Kīlauea Volcano, Hawai ‘i, Atmospheric Chemistry and Physics, 14, 8309–8322, 2014.

Businger, S., Huff, R., Pattantyus, A., Horton, K., Sutton, A. J., Elias, T., and Cherubini, T.: Observing and forecasting Vog dispersion from Kīlauea Volcano, Hawaii, Bulletin of the American Meteorological Society, 96, 1667–1686, 2015.

Kern, C., Lerner, A. H., Elias, T., Nadeau, P. A., Holland, L., Kelly, P. J., Werner, C. A., Clor, L. E., and Cappos, M.: Quantifying gas emissions associated with the 2018 rift eruption of Kīlauea Volcano using ground-based DOAS measurements, Bulletin of Volcanology, 82, 1–24, 2020.

Li, C., Krotkov, N. A., and Leonard, P.: OMI/Aura Sulfur Dioxide (SO_2) Total Column L3 1 day Best Pixel in 0.25 degree × 0.25 degree V3, https://doi.org/10.5067/Aura/OMI/DATA3008, 2020.

Pattantyus, A. K., Businger, S., and Howell, S. G.: Review of sulfur dioxide to sulfate aerosol chemistry at Kīlauea Volcano, Hawai ‘i, Atmospheric Environment, 185, 262–271, 2018.